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## (54) ADHESIVE HAVING ANISOTROPIC ELECTRICAL CONDUCTIVITY AND ADHESIVE SHEET HAVING ANISOTROPIC ELECTRICAL CONDUCTIVITY

### (57)Abstract:

**PURPOSE:** To provide a specific adhesive having anisotropic conductivity, capable of electrically connecting two substrates each having an electric connection part by the hot-pressing through the adhesive and satisfying high electrical conductivity in the pressing direction and high insulation in the non-pressed direction at the same time over a long period at time even under high temperature and humidity condition.

**CONSTITUTION:** This adhesive having anisotropic electrical conductivity is produced by dispersing electrically conductive particles in an insulating adhesive component composed of an acrylic adhesive, an epoxy resin precursor and capsules containing an imidazole-type curing agent (preferably 2-methylimidazole, 2-ethyl-4-methylimidazole, 1-cyanoethyl-2-methylimidazole, 1-cyanoethyl-2-ethyl-4-methylimidazole or 2-phenylimidazole) and having a surface layer of a resin incompatible with the imidazole compound. At least two substrates each having an electrical connection part on the surface are placed in a state to face the electrical connection parts to each other interposing the above adhesive between the connection parts and the substrates are hot-pressed to achieve the electrical connection of the electrical connection parts of the substrates in the pressing direction.

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**CLAIMS**

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[Claim(s)]

[Claim 1] By arranging and carrying out thermocompression bonding of at least two substrates with which the electrical installation section was prepared in the front face so that this electrical installation section may meet through the adhesives with which the conductive particle is distributed in the insulating adhesion component They are the anisotropic conductive adhesives which make it flow through the electrical installation section of each substrate in the pressurization direction electrically. Anisotropic conductive adhesives characterized by this insulating adhesion component consisting of acrylic adhesives, an epoxy resin precursor and an imidazole compound, and an imidazole system curing agent capsule in which the surface layer was formed with incompatible resin.

[Claim 2] Anisotropic conductive adhesives given in the 1st term of a claim with which the imidazole system curing agent contained in a capsule is characterized by being an imidazole compound meltable in water, or 2-phenylimidazole.

[Claim 3] The 1st term of a claim characterized by being at least one kind of imidazole compound chosen from the group which an imidazole system curing agent becomes from 2-methylimidazole, 2-ethyl-4-methylimidazole, 1-cyanoethyl-2-methylimidazole, 1-cyanoethyl-2-ethyl-4-methylimidazole, and 2-phenylimidazole, or anisotropic conductive adhesives given in the 2nd term.

[Claim 4] Anisotropic conductive adhesives given in the 1st term of a claim with which the resin which forms the surface layer of an imidazole system curing agent capsule is characterized by being polyvinyl alcohol.

[Claim 5] Anisotropic conductive adhesives given in the 1st term of a claim characterized by being the capsule formed by the spray-drying method which set the aqueous dispersion liquid in which an imidazole system curing agent capsule contains an imidazole compound, this imidazole and the resin which does not have compatibility, and non-subtlety powder as conditions with 100-200 degrees C [ of inlet temperatures ], and an outlet temperature of 65-95 degrees C.

[Claim 6] The anisotropic conductive adhesion sheet characterized by carrying out size enlargement of the anisotropic conductive adhesives of either the 1st term of a claim thru/or the 5th term given in a term to the shape of a sheet of 10-50-micrometer thickness.

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the anisotropic conductive adhesion sheet which carried out size enlargement of the adhesives and these adhesives for making it flow through the circuit pattern of these two substrates only in the pressurization direction electrically to the shape of a sheet by arranging at least two substrates with which the circuit pattern (electrical installation section) was prepared in the front face so that each circuit pattern may meet, and carrying out thermocompression bonding through adhesives.

[0002]

[Background of the Invention] While forming electric conductivity between the circuit patterns which meet by pasting up two or more wiring substrates with which the circuit pattern was formed in the front face in the condition of having made each wiring substrate meeting, between the circuit patterns of the same substrate, anisotropic conductive adhesives are used as adhesives for maintaining an insulating condition. These anisotropic conductive adhesives are the adhesives which distributed the conductive particle in the insulating adhesion component (refer to official reports, such as JP, 62-206772, A, 62-40183, and 62-40184). If it carries out from the function to pressurize these anisotropic conductive adhesives and to paste up two substrates, although it is a kind of a pressure sensitive adhesive, a different property from a common pressure sensitive adhesive in that each component is blended so that it may have electric conductivity only in the pressurization direction is required by the whole adhesives' having insulation and carrying out heating pressurization adhesion.

[0003] Namely, of course, in addition to it, that bond strength is high has energization nature only in the fixed direction by adhesion in a desired location so that anisotropic conductive adhesives may be required of a pressure sensitive adhesive, and the very special property of having insulation is required in other directions.

[0004] The above anisotropic conductivity carries out heating pressurization on both sides of anisotropic conductive adhesives between two wiring substrates. Both substrates are pasted up, while moving the insulating adhesives of a part with which the circuit pattern was formed to a longitudinal direction and maintaining the insulation between the circuit patterns on the same substrate. And it is the property discovered making it flow through the circuit pattern arranged at each substrate in the pressurization direction by the conductive particle, i.e., by connecting between different substrates electrically.

[0005] Various adhesive resin is used as insulating adhesives which form such anisotropic conductive adhesives. For example, invention of the anisotropic conductive

film which is formed in JP,4-95310,A from the mixture containing a reactant elastomer, an epoxy resin, the solvent that dissolves these, an imidazole derivative epoxy compound, a nonresponsive diluent, and a conductive particle, and changes is indicated.

[0006] Moreover, invention of the anisotropic conductive film which carries out the sialorrhea and desiccation of the mixed solvent which contains a reactant elastomer, an epoxy resin, the solvent that dissolves these, the microencapsulated imidazole derivative, and a conductive particle in JP,4-192212,A on a carrier film, and comes to produce a film is indicated.

[0007] Furthermore, invention of the anisotropic conductive film which carries out the sialorrhea and desiccation of the mixed solution which contains in JP,5-32799,A the reactant elastomer which mixed the silane coupling agent to homogeneity, an epoxy resin and the solvent which dissolves these, a microencapsulation imidazole derivative, and a conductive particle on a carrier film, and comes to produce a film is indicated.

[0008] In the adhesives constituent containing the above epoxy resins, although the imidazole derivative is used as a curing agent of an epoxy resin (precursor), since an imidazole and an epoxy resin precursor have the high reaction rate, in order to suppress the reactivity of an imidazole, the microcapsule of an imidazole which the imidazole and the epoxy resin precursor were made to react beforehand, and was prepared is used.

[0009] That is, in the above conventional techniques, it is consumed in order for a part of imidazole which should react as a curing agent to form a capsule and to form an epoxy resin precursor and an addition product, and the activity of the whole imidazole may fall by using a part of imidazole as a capsule ingredient in this way. For example, if the imidazole to which the part reacted with the epoxy resin precursor as mentioned above is used as a curing agent, there will be an inclination for the original adhesive property to become low, and the adhesive property will fall with the passage of time further. The adhesive fall especially in a high-humidity/temperature condition is remarkable. Such an adhesive fall appears notably as decline of the energization nature in different direction electric conduction. That is, if the anisotropic conductive adhesives which used the capsulation imidazole to which a part of imidazole was made to react to with an epoxy resin precursor etc., the capsule was formed in, and activity fell are left about one month in a high-humidity/temperature condition, they cannot maintain practical energization nature in many cases.

[0010] Furthermore, if a part of imidazole is made to react with an epoxy resin precursor and a microcapsule is formed before the above adhesion, in order for the activity of an imidazole to fall, the time amount of heating sticking by pressure becomes long, and a problem that electronic parts may heat-deteriorate with this heating is also produced.

[0011]

[Objects of the Invention] This invention aims at offering the anisotropic conductive adhesives and the sheet which are satisfied with coincidence of the good conductivity to the pressure direction, and the good insulation to the non-pressure direction over a long period of time also in the high-humidity/temperature condition.

[0012] Furthermore, this invention aims at offering the anisotropic conductive adhesives and the sheet which sufficient bond strength discovers, even if heating sticking-by-pressure time amount is short.

[0013]

[Summary of the Invention] The anisotropic conductive adhesives of this invention by

arranging and carrying out thermocompression bonding of at least two substrates with which the electrical installation section was prepared in the front face so that this electrical installation section may meet through the adhesives with which the conductive particle is distributed in the insulating adhesion component. They are the anisotropic conductive adhesives which make it flow through the electrical installation section of each substrate in the pressurization direction electrically. It is characterized by this insulating adhesion component consisting of acrylic adhesives, an epoxy resin precursor and an imidazole compound, and an imidazole system curing agent capsule in which the surface layer was formed with incompatible resin.

[0014] Moreover, the anisotropic conductive adhesion sheet of this invention is characterized by carrying out size enlargement of the above-mentioned anisotropic conductive adhesives to the thickness of 10-50 micrometers. The imidazole system curing agent capsule contained in the anisotropic conductive adhesives of this invention does not have an imidazole and reactivity, and the surface layer is formed by the resin which moreover is not compatible in this imidazole. Therefore, since this imidazole has not reacted at all before heating sticking by pressure, the activity of the used imidazole is held as it is.

[0015] And by using a meltable imidazole compound for water as this imidazole compound, especially the anisotropic conductive adhesives of this invention show the outstanding adhesive property, and even if this outstanding adhesive property leaves a pasted up object under a high-humidity/temperature condition for a long period of time, they do not fall.

[0016] Moreover, with the anisotropic conductive adhesives of this invention, since the high activity of the imidazole which is a curing agent is maintained as it is, heating sticking-by-pressure time amount can be shortened. Therefore, electronic parts do not heat-deteriorate with heating in the case of heating sticking by pressure.

[0017]

[Detailed Description of the Invention] Next, the anisotropic conductive adhesives and the sheet of this invention are explained concretely. The anisotropic conductive adhesives of this invention consist of an insulating adhesives component and conductive metal particles distributed in this insulating adhesion component.

[0018] The insulating adhesion component which forms the anisotropic conductive adhesives of this invention consists of an imidazole system curing agent capsule in which the surface layer was formed by acrylic adhesives, the epoxy resin precursor, and specific resin.

[0019] As acrylic adhesives, the adhesives of an acrylic ester (meta) system are used preferably here. As an example of this acrylic resinous principle, the copolymer of acrylic ester (meta), and a this and the compound which has a copolymerizable reactant double bond can be mentioned. As an example of the acrylic ester used here (meta) Methyl (meta) acrylate, ethyl (meta) acrylate, isopropyl (meta) acrylate, Butyl (meta) acrylate, 2-ethylhexyl (meta) acrylate, Lauryl (meta) acrylate, stearyl (meta) acrylate, Cyclohexyl (meta) acrylate, 2-hydroxyethyl (meta) acrylate, 2-hydroxypropyl (meta) acrylate, chloro-2-hydroxypropyl (meta) acrylate, Diethylene-glycol monochrome (meta) acrylate, methoxy ethyl (meta) acrylate, ethoxyethyl (meta) acrylate, dimethylaminoethyl (meta) acrylate, and glycidyl (meta) acrylate can be mentioned.

[0020] As an example of the compound which has the above (meta) acrylic ester and a

copolymerizable reactant double bond, an unsaturated-carboxylic-acid monomer, a styrene system monomer, a vinyl system monomer, etc. can be mentioned.

[0021] As an example of an unsaturated-carboxylic-acid monomer, addition polymerization nature partial saturation aliphatic series dicarboxylic acid, such as addition polymerization nature partial saturation aliphatic series monocarboxylic acid; maleic acids, such as an acrylic acid, methacrylic acid, alpha-ethyl acrylic acid, a crotonic acid, alpha-methyl crotonic acid, alpha-ethyl crotonic acid, isocrotonic acid, tiglic acid, and a UNGERIKA acid, a fumaric acid, an itaconic acid, a citraconic acid, mesaconic acid, glutaconic acid, and dihydro muconic acid, can be mentioned here.

[0022] moreover -- as the example of a styrene system monomer -- halogenation styrene [ , such as alkyl styrene; FURORO styrene, such as styrene, methyl styrene, dimethyl styrene, trimethyl styrene ethyl styrene, diethyl styrene, triethyl styrene, propyl styrene, butyl styrene, hexyl styrene, heptyl styrene, and octyl styrene, chloro styrene, bromostyrene, dibromo styrene, and iodine styrene, ]; -- nitro styrene, acetyl styrene, and methoxy styrene can be mentioned further.

[0023] Furthermore, as an example of a vinyl system monomer, halogenation vinylidenes [ , such as a halogenation vinyl; vinylidene chloride, ], such as a conjugated diene monomer; vinyl chloride, vinyl bromide, etc., such as vinylpyridine, vinyl pyrrolidone, vinylcarbazole, a divinylbenzene, vinyl acetate and an acrylonitrile; butadiene, an isoprene, and a chloroprene, can be mentioned.

[0024] In 60 - 90 weight section and monomers other than this, copolymerization of the above-mentioned acrylic ester (meta) is usually carried out in the amount of 10 - 40 weight section, and acrylic adhesives are usually manufactured [ acrylic ester ].

[0025] Such acrylic adhesives can be manufactured by the usual approach. For example, an organic solvent can be made to be able to dissolve or distribute the above-mentioned monomer, and it can manufacture by making these solution or dispersion liquid react in a reactor like nitrogen gas by which inerting was carried out. As an example of the organic solvent used here, ketones, such as fatty alcohol, such as ester, such as aliphatic hydrocarbon, such as aromatic hydrocarbon, such as toluene and a xylene, and n-hexane, ethyl acetate, and butyl acetate, n-propyl alcohol, and i-propyl alcohol, a methyl ethyl ketone, methyl isobutyl ketone, and a cyclohexanone, can be mentioned. An organic solvent is usually used in the amount of the 100 - 250 weight section to the acrylic adhesives formation raw material 100 weight section by the above-mentioned reaction.

[0026] This reaction is performed by heating under existence of a polymerization initiator. As an example of the reaction initiator used here, azobisisobutyronitril, benzoyl peroxide, G tert-butyl peroxide, a cumene hydroperoxide, etc. can be mentioned. This polymerization initiator is usually used in the amount of 0.01 - 5 weight section to the acrylic adhesives formation raw material 100 weight section.

[0027] The polymerization reaction in the above organic solvents usually heats reaction mixture at 60-75 degrees C, and is usually performed preferably for 4 to 8 hours for 2 to 10 hours. In this way, although the manufactured acrylic resin can also be used dissociating from a reaction solvent, it is desirable to use the generated resin, where an organic solvent is dissolved or distributed.

[0028] For example, about the acrylic adhesives manufactured as mentioned above, the elastic modulus measured at the temperature of 200 degrees C is usually within the limits of 105 - 107 dyn/cm<sup>2</sup>. The insulating adhesives component which forms the anisotropic

conductive adhesives of this invention contains the epoxy resin precursor. It is what this epoxy resin precursor reacts with a curing agent, and forms an epoxy resin hardening object. As an epoxy resin precursor Bisphenol A, the epoxy resin precursor of an epichlorohydrin mold, ethylene glycol diglycidyl ether, Polyethylene glycol diglycidyl ether, glycerol diglycidyl ether, Glycerol triglycidyl ether, 1, 6-hexanediol diglycidyl ether, A trimethylolpropane-triglycidyl-ether, diglycidyl aniline, diglycidyl amine, N and N, N', N'-tetraglycidyl ether-meta xylene diamine and 1, and 3-bis(N and N'-diglycidyl aminomethyl) cyclohexane can be mentioned.

[0029] such an epoxy resin precursor -- the above-mentioned acrylic adhesives 100 weight section -- receiving -- 10 - 60 weight section -- it is preferably used in the amount of 30 - 50 weight section. The insulating adhesives component which forms the anisotropic conductive adhesives of this invention contains the imidazole system curing agent capsule in which the surface layer was formed with specific resin.

[0030] While being able to shorten the heating sticking-by-pressure time amount using anisotropic conductive adhesives so that the activity of the imidazole compound which an imidazole compound is a curing agent to the above-mentioned epoxy resin precursor, and is used is high in this invention, initial bond strength can be made high. The adhesion property furthermore discovered in early stages is maintainable for a long period of time. However, since an imidazole will react for a short time if it contacts an epoxy resin precursor in the condition as it is The \*\* to which a reaction does not advance even if it adds an epoxy resin precursor to some imidazole compounds used as a curing agent, it forms an imidazole capsule and an imidazole capsule and an epoxy resin precursor contact in ordinary temperature conventionally, A capsule is destroyed and he was trying for an imidazole and an epoxy resin precursor to react from passing through predetermined actuation of heating etc.

[0031] In the anisotropic conductive adhesives of this invention, the surface layer is formed by incompatible resin with this imidazole compound, without an imidazole system curing agent capsule making the imidazole compound which is a curing agent like before completely involve as a raw material which forms the surface layer of a capsule. Therefore, the imidazole compound can be effectively used as a curing agent of an epoxy resin precursor.

[0032] Since it is not participating in formation of the surface layer of a capsule directly, the imidazole compound used by this invention can be chosen mainly in consideration of labile with an epoxy resin precursor.

[0033] As an imidazole compound which can be used by this invention, to an organic solvent (for example, toluene, ethyl acetate) like 2-phenylimidazole, 1-benzyl-2-methylimidazole, 2-undecylimidazole, 2-pentadecylimidazole, 1-cyanoethyl-2-phenylimidazole, and 1-cyanoethyl-2-undecylimidazole, it is meltable and a refractory imidazole can be mentioned to water. The imidazole refractory in such water has the hydrocarbon groups (example: a phenyl group and an aromatic hydrocarbon radical like benzyl, an undecyl radical, an aliphatic hydrocarbon radical like a pentadecyl group, etc.) of at least one carbon atomic number [ six or more ] in the location of either the 1st place of an imidazole ring, the 2nd place or the 4th place. By such existence of a radical with a comparatively big carbon number, these imidazole compounds become poor solubility to water, and become fusibility to an organic solvent. And the reaction of this imidazole compound and an epoxy resin precursor becomes comparatively mild by such existence

of a radical with a comparatively big carbon number. However, as compared with an imidazole refractory in other water, labile of 2-phenylimidazole is remarkably high in an imidazole refractory in the above water.

[0034] Furthermore, as an imidazole compound which can be used by this invention, a meltable imidazole can be mentioned to water like 2-methylimidazole, 2-ethyl-4-methylimidazole, 1-cyanoethyl-2-methylimidazole, and 1-cyanoethyl-2-ethyl-4-methylimidazole. The carbon atomic number of the hydrocarbon group which is a substituent which has an imidazole meltable in this water in the location of the 1st place of an imidazole ring, the 2nd place, or the 4th place is two (example: a methyl group, ethyl group) or less preferably five or less, and the property of the water solubility which the imidazole essentially has by existence of these substituents is not impaired so much. And since the carbon atomic number of a substituent is small in this way, there is almost no fall of the labile by the steric hindrance resulting from these radicals etc., therefore it has the property that an imidazole meltable in this water has the high reactivity over an epoxy resin precursor as compared with an imidazole (except for 2-phenylimidazole) refractory in the above-mentioned water.

[0035] In addition, it means an imidazole 0.1g or more dissolving soluble completely in water at 100ml of 25-degree C water, and forming a transparent water solution at it here, and means that insoluble matter remains without an imidazole dissolving completely also by supplying a 0.1g imidazole at 100ml of 25-degree C water as it is refractory, and stirring in water.

[0036] Although both an imidazole refractory in the above water and an imidazole meltable in water can be used in this invention, when a refractory imidazole is used for the water except 2-phenylimidazole, it is necessary to make longer than the case where a meltable imidazole is used for water some heating time which adhesion takes.

[0037] The surface layer which becomes the front face of a drop which consists of the above imidazoles from the resin which is not compatible in this imidazole is formed in the imidazole system curing agent capsule used by this invention. And since the resin which forms this surface layer does not have the reactivity over an imidazole, either, the chemical integrated state is not formed between the surface layer which consists of such resin, and the imidazole enclosed with this interior.

[0038] It is resin which does not have compatibility to an imidazole as resin which forms the surface layer of such a capsule, and does not have reactivity to an imidazole, and meltable resin can be further used for water.

[0039] As an example of such resin, polyvinyl alcohol, starch, gelatin, poly acrylamide, polyethylene oxide, a polyvinyl pyrrolidone, and polyvinyl ether can be mentioned. In this invention, polyvinyl alcohol is especially desirable. The surface layer formed with such resin is usually about 2 micrometers that the epoxy resin precursor which is an insulating adhesives nature component, and the imidazole enclosed into the capsule can be divided in the case of preservation or migration, and both should just have the thickness which is promptly mixable extent in the case of heating sticking by pressure. Moreover, the mean particle diameter of this imidazole system curing agent capsule is usually within the limits of 5-300 micrometers. Homogeneous high adhesives can be formed by using the imidazole system curing agent capsule which has the above mean particle diameter.

[0040] Although such an imidazole system curing agent capsule can be manufactured by



various approaches, it is desirable especially to manufacture by the spray-drying method by this invention. This spray-drying method makes an aqueous medium distribute this non-subtle powder, when non-subtle powder is blended, while dissolving the resin which forms an imidazole and a surface layer, and the resin which forms an imidazole and a surface layer by feeding non-subtle powder into an aqueous medium as occasion demands further, and carrying out stirring etc. in this aqueous medium.

[0041] As non-subtle powder blended by the need here, 0.01-5 micrometers of mean particle diameter are usually within the limits of 0.05-3 micrometers preferably, and the inorganic particle which has 1 / about ten to 1/100 mean particle diameter of the mean particle diameter of an imidazole system curing agent capsule obtained is used. As an example of such non-subtle powder, a silica, titanium oxide, magnesium oxide, a zirconium dioxide, a calcium carbonate, a magnesium carbonate, calcium phosphate, boron nitride, silicon nitride, boron aluminum, and an antimony trioxide can be mentioned. By blending such non-subtle powder, the stability of the surface layer of an imidazole system curing agent capsule with the passage of time improves.

[0042] the inside of the aqueous solution 100 weight section used in order to form a capsule by the spray-drying method -- an imidazole compound -- usually -- 0.1 - 10 weight section and the resin which forms 2 - 5 weight section and a surface layer preferably -- usually -- 0.1 - 10 weight section -- preferably, 2 - 5 weight section and when using non-subtle powder further, each component is blended so that it may usually contain in the amount of 0.1 - 2 weight section.

[0043] The spray conditions for forming a capsule from the above aqueous solutions have an inlet temperature set up as 150-200 degrees C, and 100-200 degrees C of outlet temperature are usually preferably set up within the limits of 65-95 degrees C. On the above temperature conditions, the flow rate of an aqueous solution is adjusted so that the spray dry processing of the 1200g aqueous solution can usually be preferably carried out in 60 - 100 minutes for 60 to 180 minutes. Moreover, it is desirable to use 2 hydraulic nozzles on the occasion of this spray dry, and 1.5-3kg/cm<sup>2</sup> of pressures at this time is usually preferably set as the range of 1.5-3kg/cm<sup>2</sup>.

[0044] By spray-drying as mentioned above, the imidazole system curing agent capsule whose mean particle diameter is about 5-300 micrometers can be manufactured. The surface layer which consists of resin which this imidazole system curing agent capsule does not dissolve in an imidazole like polyvinyl alcohol on a front face is formed, and the imidazole compound is enclosed with this interior. And chemical bonding strength does not exist between the resin which forms this surface layer, and the imidazole compound enclosed with the interior.

[0045] As for the imidazole compound with which it is used by this invention since the imidazole system curing agent capsule used by this invention as mentioned above is formed by dissolving an imidazole compound and a capsule shell formation ingredient in an aqueous medium suitably, and spray-drying this solution, it is desirable that it is an imidazole meltable in water. Furthermore, when it compares about reactivity with the epoxy resin precursor of an imidazole meltable in water, and an imidazole refractory in water, activity is higher than what has the one of an imidazole meltable in water refractory in water. In addition, although 2-phenylimidazole is refractory in water, if it is examined in a detail about reactivity with an epoxy resin precursor, it has labile equivalent to 2-methylimidazole etc., and has the heterogeneous property in the

refractory imidazole in other water.

[0046] Thus, by using a meltable imidazole or 2-phenylimidazole for the water which carried out [ above-mentioned ] instantiation, the heating sticking-by-pressure time amount which adhesion takes can be shortened. When a refractory imidazole (except for 2-phenylimidazole) is used for water, in order to obtain sufficient initial bond strength, when a meltable imidazole or 2-phenylimidazole is used for water to it being common that the sticking-by-pressure temperature of 180 degrees C and conditions with a pressure of 45kg/cm<sup>2</sup> take the heating time for 30 seconds or more, this heating time can be shortened at about 5 seconds.

[0047] Thus, an imidazole compound meltable in using what formed the imidazole system curing agent capsule by the spray-drying method suitably in this invention, and water has activity higher than an imidazole compound refractory in water, and it is desirable to use a meltable imidazole compound for the above-mentioned water as this imidazole compound from the reasons of being able to shorten pressurization sticking-by-pressure time amount remarkably. Furthermore, if reactivity with an epoxy resin precursor is taken into consideration, in addition to a meltable imidazole compound, 2-phenylimidazole can also be further used for the above-mentioned water preferably.

[0048] And even if it uses the imidazole of high activity in this way, an epoxy resin precursor and an imidazole system hardening object capsule can be made to live together in the condition of having been stabilized, by using the resin which has neither the reactivity over an imidazole like polyvinyl alcohol, nor compatibility as resin for surface formation.

[0049] although the amount used changes with the class of imidazole compound which uses such an imidazole system curing agent capsule, classes of epoxy resin precursor, etc. -- the above-mentioned epoxy resin precursor 100 weight section -- receiving -- 1 - 20 weight section -- it is preferably used in the amount of 1 - 10 weight section. By blending an imidazole system curing agent capsule in the above amount, since the surface layer formed from polyvinyl alcohol etc. is destroyed, an imidazole and an epoxy resin precursor contact, it reacts promptly, the network of an epoxy resin hardening object is formed into acrylic adhesives and a flow of acrylic adhesives with time is restricted by formation of such a network at the time of heating sticking by pressure, the moisture-proof thermal resistance of the anisotropic conductive adhesives of this invention is secured.

[0050] The conductive particle is distributed in the above insulating adhesion components by the anisotropic conductive adhesives of this invention. There are conductive metal particles, and a conductive inorganic material particle and the composite particle which covered the front face of a resin core material with the conductive ingredient in the conductive particle used here. The component which forms the conductive metal particles, the conductive inorganic material particle, or the conductive ingredient which forms a conductive particle is the alloy which usually contains 100ohms or less of the conductive metals in which the electric resistance value of 10 - 1 or less ohm is shown preferably and these metals, a conductive ceramic, conductive metallic oxide, or other conductive ingredients.

[0051] As an example of a conductive metal, Zn, aluminum, Sb, U, Cd, Ga, calcium, Au, Ag, Co, Sn, Se, Fe, Cu, Th, Pb, nickel, Pd, Be, and Mg can be mentioned. Moreover, the above-mentioned metal may be used independently, may use two or more kinds, and may

add the element of further others, a compound (for example, pewter), etc. As an example of the conductive ceramic which is a conductive inorganic material, VO<sub>2</sub>, Ru<sub>2</sub>O, SiC and ZrO<sub>2</sub>, Ta<sub>2</sub>N, ZrN, NbN, VN, TiB<sub>2</sub>, ZrB, HfB<sub>2</sub>, TaB<sub>2</sub>, MoB<sub>2</sub> and CrB<sub>2</sub>, and B<sub>4</sub>C, MoB, ZrC, VC, and TiC can be mentioned. Moreover, carbon, a carbon particle like graphite, ITO, etc. can be mentioned as conductive ingredients other than the above.

[0052] Conductive metal particles or a conductive inorganic material particle is a particle which consists of the above components, and usual has still more preferably 2-20 micrometers of 1-48 micrometers of the mean particle diameter within the limits of 5-10 micrometers preferably.

[0053] Moreover, the composite particle used as a conductive particle by this invention is a particle which covered the front face of glass and an inorganic core material like an alumina, or a resin core material with the above conductive ingredients.

[0054] As an example of the resin which thermosetting resin or thermoplastics may be formed either and forms a core material, the resin core material used here Phenol resin, a urea resin, melamine resin, allylic resin, furan resin, Polyester resin, an epoxy resin, silicone resin, polyamidoimide resin, polyimide resin, polyurethane resin, a fluororesin, and polyolefin resin (example: -- polyethylene --) Polypropylene, polybutylene, polyalkyl (meta) acrylate resin, Pori (meta) acrylic acid resin, polystyrene resin, acrylonitrile-styrene-butadiene resins, Vinyl resin, polyamide resin, polycarbonate resin, polyacetal resin, Ionomer resin, polyether sulphone resin, polyphenyl oxide resin, polysulfone resin, polyvinylidene fluoride resin, ethyl cellulose, and cellulose acetate can be mentioned.

[0055] The core material formed from an inorganic core material or the above resin 1-48 micrometers usually has still more preferably 2-20 micrometers of mean particle diameter of 5-10 micrometers preferably.

[0056] The conductive layer currently formed in the front face of the above-mentioned core material is formed from the alloy containing conductive metals and these metals, a conductive ceramic, conductive metallic oxide, or other conductive ingredients.

[0057] As an example of a conductive metal, Zn, aluminum, Sb, U, Cd, Ga, calcium, Au, Ag, Co, Sn, Se, Fe, Cu, Th, Pb, nickel, Pd, Be, and Mg can be mentioned. Moreover, the above-mentioned metal may be used independently, may use two or more kinds, and may add the element of further others, a compound (for example, pewter), etc. As an example of a conductive ceramic, VO<sub>2</sub>, Ru<sub>2</sub>O, SiC and ZrO<sub>2</sub>, Ta<sub>2</sub>N, ZrN, NbN, VN, TiB<sub>2</sub>, ZrB, HfB<sub>2</sub>, TaB<sub>2</sub>, MoB<sub>2</sub> and CrB<sub>2</sub>, and B<sub>4</sub>C, MoB, ZrC, VC, and TiC can be mentioned.

Moreover, carbon, a carbon particle like graphite, ITO, etc. can be mentioned as conductive ingredients other than the above.

[0058] Such a conductive layer Vacuum deposition, the ion sputtering method, plating, Physical methods, such as a spraying process, the chemical approach of combining a conductive ingredient with the resin core material front face which has a functional group chemically, It can form by the approach of forming a core material and a conductive layer in coincidence etc., making a conductive particle live together in the system of reaction, and depositing a conductive particle on the surface of a core material, in case the approach and core material to which a conductive ingredient is made to stick on the surface of a core material using a surfactant etc. are formed. It is desirable to form this conductive layer especially by the electroless deposition method. Such a conductive layer does not need to be a monolayer and the laminating of two or more layers may be carried out.

[0059] Usual has still more preferably 0.05-5 micrometers of 0.01-10.0 micrometers of thickness of such a conductive layer within the limits of 0.2-2 micrometers preferably. The insulating layer which consists of insulating resin further may be formed in the front face of this composite particle. It has covered with the usual condition (natural condition) of not applying a pressure etc. so that a conductive ingredient may not expose the front face of a conductive particle, but this insulating layer is formed so that heating and pressurization at the time of pasting up two substrates using anisotropic conductive adhesives may break and a conductive layer may be exposed. This insulating layer is formed from stable resin etc. to the solvent and adhesives component which are dissolving or distributing the insulating adhesion component. As an example of such resin, phenol resin, a urea resin, melamine resin, Allylic resin, furan resin, polyester resin, an epoxy resin, silicone resin, Polyamidoimide resin, polyimide resin, polyurethane resin, a fluororesin, Polyolefin resin (example: polyethylene, polypropylene, polybutylene), Poly alkyl (meta) acrylate resin, Pori (meta) acrylic acid resin, Polystyrene resin, acrylonitrile-styrene-butadiene resins, Vinyl resin, polyamide resin, polycarbonate resin, polyacetal resin, Ionomer resin, polyether sulphone resin, polyphenyl oxide resin, polysulfone resin, polyvinylidene fluoride resin, ethyl cellulose, and cellulose acetate can be mentioned. It is desirable to form especially this insulating layer from polyvinylidene fluoride resin.

[0060] As an approach of forming an insulating layer in a conductive layer front face, the physicochemical approaches, such as the physical mechanical approaches, such as the chemical approaches, such as the coacervation method, interfacial polymerization, an insitu polymerization method, and hardening-among liquid coating, a spray drying process, suspension-among mind coating, vacuum deposition coating, the dryblend method, the electrostatic coalescing method, a fusion distribution cooling method, and minerals encapsulation, and an interface precipitation method, can be used. Also in these, the physical mechanical approach is desirable and especially the dryblend method (example: the covering approach using a hybridization system) is still more desirable.

[0061] If the example of the approach of forming the discontinuous insulating layer which consists of polyvinylidene fluoride for example, by the hybridization system as an approach of forming an insulating layer is shown, it will process for 5 - 10 minutes at the temperature of 85-115 degrees C using the polyvinylidene fluoride of 2 - 8 weight section to the conductive particle 100 weight section.

[0062] In addition, even when using arts other than a hybridization system, an insulating layer can be formed by changing the processing condition according to the above-mentioned case.

[0063] This insulating layer thickness is usually about 0.1-0.5 micrometers. In addition, this insulating layer may cover the front face of a conductive particle imperfectly. such a conductive particle -- the insulating adhesives (resin weight) 100 weight section -- receiving -- usually -- the 5 - 100 weight section -- it is preferably blended in the amount of 20 - 60 weight section.

[0064] Furthermore, it is desirable to blend a filler into the adhesives used by this invention. An insulating inorganic particle can be used as a filler here, and titanium oxide, a silicon dioxide, a calcium carbonate, calcium phosphate, an aluminum oxide, and an antimony trioxide can be mentioned as this example. This insulating inorganic particle usually has the mean particle diameter of 0.01-5 micrometers. this insulating inorganic

particle is independent -- it is -- it can be combined and used.

[0065] this insulating inorganic particle -- the resinous principle 100 weight section in adhesives -- receiving -- usually -- the 1.0 - 50.0 weight section -- it is preferably used in the amount of the 5.0 - 25.0 weight section. By blending such an insulating inorganic particle in the above-mentioned amount as a filler, even if it can adjust the fluidity of adhesives and heats after adhesion, adhesives flowing backwards and checking conductivity decreases. Moreover, the flash of the adhesives from the edge of a printed circuit board can be prevented in the case of adhesion.

[0066] The anisotropic conductive adhesives of this invention can be manufactured by mixing each above-mentioned component. The anisotropic conductive adhesives of this invention can be used with various gestalten, such as the shape of the shape of a sheet (the shape of a film), and a paste.

[0067] Since the imidazole compound which is a curing agent is encapsulated and stabilized, as for especially the anisotropic conductive adhesives of this invention, it is desirable to use it by making it the shape of a sheet by making the solvent which does not corrode the surface layer of an imidazole system curing agent capsule distribute acrylic adhesives, an epoxy resin precursor, an imidazole system curing agent capsule, and a conductive particle, applying these dispersion liquid, and removing a dispersion medium. And when making it the shape of such a sheet, it is desirable to carry out thickness of this sheet within the limits of 10-50 micrometers.

[0068] As an example of the dissolution or the solvent which can be distributed good, toluene, ethyl acetate, butyl carbitol, and acetate can be mentioned for other components, without affecting the surface layer of the imidazole system curing agent capsule used in case a sheet is formed here.

[0069] For example, in order to make the anisotropic conductive adhesives of this invention into the shape of a sheet, a knife coating machine, a comma coating machine, a reverse roll coater, a gravure coating machine, etc. can be used.

[0070] As the anisotropic conductive adhesion sheet of this invention which carried out size enlargement to the shape of a sheet is shown in drawing 1, it can be used for it. The adhesion approach of the substrate using the anisotropic conductive adhesives of this invention made into the shape of a sheet at drawing 1 is shown typically.

[0071] As shown in [a] of drawing 1, two substrates 18a and 18b with which circuit patterns 19a and 19b were formed in the front face are arranged so that circuit patterns 19a and 19b may meet in the meantime, and the anisotropic conductive adhesives 17 (anisotropic conductive adhesion sheet) of this invention fabricated in the shape of a sheet among these circuit patterns 19a and 19b are put. The particle 15 and filler 16 by which the insulating layer was formed in the particle front face which consists of a conductive ingredient into the insulating adhesives 21 with which this anisotropic conductive adhesion sheet 17 consists of acrylic adhesives, an epoxy resin precursor, and an imidazole system curing agent capsule are distributed.

[0072] Thus, if it pressurizes and the bottom of heating is pasted in the direction of an arrow head shown in [a], the substrates 18a and 18b with which the anisotropic conductive adhesion sheet 17 has been arranged The particle 15 among circuit patterns 19a and 19b receives the highest pressure. It is destroyed, the insulating layer formed in the front face of this particle 15 is extruded by the part in which a circuit pattern is not formed, i.e., the pressurization direction, and the longitudinal direction of a right angle

with an excessive adhesives component, and circuit pattern 19a and circuit pattern 19b flow. And the insulating layer currently formed on the surface of the particle is destroyed by the pressurization under such heating. Therefore, an insulating layer is easily removed from the front face of the particle in the part pinched with the circuit pattern, and such particle 15a comes to have good conductivity. On the other hand, since such a pressure is not applied to particle 15b in the part in which the circuit pattern is not formed, good insulation is shown.

[0073] And by carrying out pressurization sticking by pressure in this way, the surface layer of the imidazole system curing agent capsule contained in the insulating adhesives 21 is destroyed by this pressure and heat, an imidazole and an epoxy resin precursor contact, and an epoxy resin is formed. Although the above showed the mode which uses it by making the anisotropic conductive adhesives of this invention into the shape of a sheet, when the anisotropic conductive adhesives of this invention contain a suitable solvent, it can also be used by the shape of a paste. The anisotropic conductive adhesives of the shape of this paste can be applied on a substrate for example, using a screen coating machine etc., and can be used like the above.

[0074]

[Effect of the Invention] The imidazole system curing agent capsule contained in the anisotropic conductive adhesives of this invention does not have an imidazole and reactivity, and the surface layer is formed by the resin which moreover is not compatible in this imidazole. Therefore, since this imidazole has not reacted at all before heating sticking by pressure, the activity of the used imidazole is held as it is.

[0075] And by using a meltable imidazole compound for water as this imidazole compound, especially the anisotropic conductive adhesives of this invention show the outstanding adhesive property, and even if this outstanding adhesive property leaves a pasted up object under a high-humidity/temperature condition for a long period of time, they do not fall.

[0076] Moreover, since the high activity of the imidazole which is a curing agent is maintained as it is, the anisotropic conductive adhesives of this invention can shorten heating sticking-by-pressure time amount. Therefore, electronic parts do not heat-deteriorate with heating in the case of heating sticking by pressure.

[0077] Furthermore, since the reaction of an epoxy resin precursor and an imidazole does not advance during storage even if it makes it a liquid type and carries out size enlargement to the shape of a sheet, since the storage stability of the imidazole compound which is a curing agent is high, the anisotropic conductive adhesion sheet of this invention can use this sheet for stability for a long period of time.

[0078]

[Example] Next, although an example is shown and the anisotropic conductive adhesives of this invention are explained, this invention is not restrictively interpreted according to these examples.

[0079] In addition, in this invention, the coverage of initial conductivity, moisture-proof conductivity, insulation, an adhesive property, and a particle was measured by the approach of indicating below. Heating a polyimide film with a thickness of 50 micrometers which has arranged copper foil in pitch of 200 micrometers, and the glass plate which carried out sputtering of the ITO to the pitch of 200 micrometers on both sides of an anisotropic conductive adhesion sheet with a thickness of 25 micrometers to

the temperature of 150 degrees C, 180 degrees C, or 210 degrees C, as shown in drawing 2, it pressurized by the predetermined pressure and the polyimide film and the glass plate were pasted up.

[0080] The resistance after the bottom of 24-hour neglect and between a vertical electrode (initial conductivity), moisture-proof backward conductivity (moisture-proof conductivity), inter-electrode insulation (insulation) on either side, and the 90-degree bond strength (adhesive property) in 10mm width of face were measured for this at 23 degrees C.

[0081] In addition, moisture-proof conductivity was measured after leaving a sample for 14 days on condition that 60 degrees C and 90%RH.

[0082]

[Example 1]

The xylene 200 weight section, the methyl methacrylate 70 weight section, the butyl acrylate 25 weight section, and the acrylic-acid 5 weight section were taught to the glass reaction container equipped with the manufacture agitator, the thermometer, nitrogen gas entrainment opening, and the reflux cooling system of adhesives.

[0083] The azobisisobutyronitril 1 weight section was added to this liquid, and stirring was started. Subsequently, after it introduced nitrogen gas and nitrogen gas permuted the air in a reactor in this glass reactor, reaction mixture was heated at 66 degrees C, the polymerization was performed for 10 hours, and the acrylic adhesion component 286 weight section was obtained (resin concentration: 33.3%).

[0084] The epoxy resin 50 weight section was added to this acrylic adhesion component 100 weight section (resinous principle: 33.3 weight sections), it mixed, and insulating adhesives were obtained.

The nickel particle whose manufacture mean particle diameter of a conductive particle is 10.2 micrometers was prepared, the polystyrene resin 4 weight section was blended with this nickel particle 100 weight section, and the particle 101 weight section by which the insulating layer which consists of polystyrene resin was formed on the nickel particle was obtained by introducing this into a hybridization system and processing for 2.5 minutes at the temperature of 85 degrees C.

[0085] The manufacture 2-phenylimidazole 30 weight section of an imidazole capsule was mixed and stirred in the water 920 weight section (A liquid).

[0086] The polyvinyl alcohol 20 weight section was independently added in the water 500 weight section, and mixed stirring was carried out (B liquid). Mixed stirring of the above-mentioned A liquid, B liquid, and silica powder (mean particle diameter: 0.05 micrometers) 2 weight section was carried out.

[0087] 2 hydraulic nozzles adjusted to the pressure of 2 kg/cm<sup>2</sup> in the obtained mixed liquor in the spray tower with 150 degrees C [ of inlet temperatures ] and an outlet temperature of 70 degrees C were used, spray drying was carried out on the conditions for the flow rate of 1400g / 120 minutes, and the imidazole capsule 47 weight section of the shape of 10-300-micrometer powder was obtained. The content of the 2-phenylimidazole in this imidazole capsule was 57.7 % of the weight.

[0088] It mixed by the presentation which shows below the adhesives which are the manufacture above of an anisotropic conductive adhesion sheet, and were made and manufactured, a conductive particle, an imidazole capsule, and silica powder (mean particle diameter: 1 micrometer), spreading desiccation of this mixture was carried out,

and the anisotropic conductive adhesion sheet with a thickness of 25 micrometers was manufactured.

[0089]

A styrene coated particle ..... Nine weight sections adhesives ..... A 100 weight sections imidazole capsule ..... 2.3 weight sections silica powder ..... It is stuck by pressure on the conditions which show the sample which kept the initial conductivity (it measures 24 hours after at 23 degrees C after sticking by pressure by the conditions shown in Table 1) and this sheet immediately after manufacture for 30 days at 40 degrees C about 20 weight \*\*\*\*\* anisotropic conductive adhesion sheet in Table 1 like the first stage in 23-degree-C ambient atmosphere. At 23 degrees C The conductivity of 24 hours after, Insulation and \*\*\*\*\* were measured. The result is shown in Table 1.

[0090]

[The example 1 of a comparison] In the example 1, the 2-phenylimidazole 1.3 weight section was blended as it was, without using an imidazole capsule, and the sheet with a thickness of 25 micrometers was similarly manufactured after this 2-phenylimidazole combination except having applied this compound immediately.

[0091] It was stuck by pressure on the conditions which show the sample which kept the initial conductivity (it measures 24 hours after at 23 degrees C after sticking by pressure by the conditions shown in Table 1) and this sheet immediately after manufacture for 30 days at 40 degrees C about the obtained anisotropic conductive adhesion sheet in Table 1 like the first stage in 23-degree-C ambient atmosphere, and the conductivity of 24 hours after, insulation, and bond strength were measured at 23 degrees C. The result is shown in Table 1.

[0092]

[Table 1]

表 1

圧着条件		導電性 (Ω)				絶縁性 (Ω)		接着性 (g/10mm)	
		初期導電性 (Ω)		30日後の導電性 (Ω)		初期絶縁性	30日後の絶縁性	初期接着性	30日後の接着性
		基板中央	基板端部	基板中央	基板端部				
150℃ 30kg/cm <sup>2</sup> 5秒	実施例 1	5 ~ 6	4 ~ 5	5 ~ 6	5 ~ 6	1 0 <sup>12</sup> 以上	1 0 <sup>12</sup> 以上	8 9 0	6 9 0
	比較例 1	5 ~ 6	5 ~ 6	1000以上	1000以上	1 0 <sup>12</sup> 以上	1 0 <sup>10</sup> 以上	6 8 0	1 0 以下
210℃ 30kg/cm <sup>2</sup> 5秒	実施例 1	4 ~ 5	4 ~ 5	4 ~ 5	5 ~ 6	1 0 <sup>12</sup> 以上	1 0 <sup>12</sup> 以上	7 1 0	7 0 0
	比較例 1	5 ~ 6	5 ~ 6	1000以上	1000以上	1 0 <sup>12</sup> 以上	1 0 <sup>9</sup> 以上	7 1 0	1 0 以下

註) 基板中央における導電性は、図 3 に示すように幅 3 0 . 0 mm の基板表面に 2 0 0 μ m 間隔で 1 5 0 本の配線パターン (ピン) が形成された基板の中心から 1 0 ~ 2 0 ピンにおける導電性を測定したものであり、端部の導電性は、図 3 に示すようにそれぞれのピン端部から 1 0 ~ 2 0 ピンにおける導電性を測定したものである。

[0093]

[Examples 2-5] In the example 1, the imidazole to be used was changed as follows, the imidazole capsule (imidazole system curing agent capsule) was manufactured, and the anisotropic conductive adhesion sheet was similarly manufactured except having used the imidazole capsule obtained instead of the imidazole capsule of this 2-phenylimidazole.

[0094] Example [ ... 1-cyanoethyl-2-ethyl-4-methylimidazole. ] 2 ... 2-methylimidazole,



example 3 ... 2-ethyl-4-methylimidazole, example 4 ... 1-cyanoethyl-2-methylimidazole, example 5

[0095] Initial conductivity (23 degrees C and 24 hours after) and the conductivity of 30 days after (40 degrees C and 30 days after) were measured about the obtained anisotropic conductive adhesion sheet. A result is shown in Table 2.

[0096]

[Examples 6-10] In the example 1, the imidazole to be used was changed as follows, the imidazole capsule (imidazole system curing agent capsule) was manufactured, and the anisotropic conductive adhesion sheet was similarly manufactured except having used the imidazole capsule obtained instead of the imidazole capsule of this 2-phenylimidazole.

[0097]

Example [ ... 1-cyanoethyl-2-phenylimidazole, example 10 / ... 1-cyanoethyl-2-undecylimidazole. ] 6 ... 1-benzyl-2-methylimidazole, example 7 ... 2-undecylimidazole, example 8 ... 2-pentadecylimidazole, example 9

[0098] Initial conductivity (23 degrees C and 24 hours after) and the conductivity of 30 days after (40 degrees C and 30 days after) were measured about the obtained anisotropic conductive adhesion sheet. A result is shown in Table 2.

[0099]

[The example 2 of a comparison] In the example 1, the anisotropic conductive adhesion sheet was similarly manufactured except having changed the imidazole capsule which connotes 2-phenylimidazole into the microencapsulation imidazole derivative which is the addition product of 2-phenyl-4-methyl-5-hydroxymethylimidazole and a bisphenol mold epoxy resin.

[0100] Initial conductivity (23 degrees C and 24 hours after) and the conductivity of 30 days after (40 degrees C and 30 days after) were measured about the obtained anisotropic conductive adhesion sheet. A result is shown in Table 2.

[0101]

[Table 2]

表 2

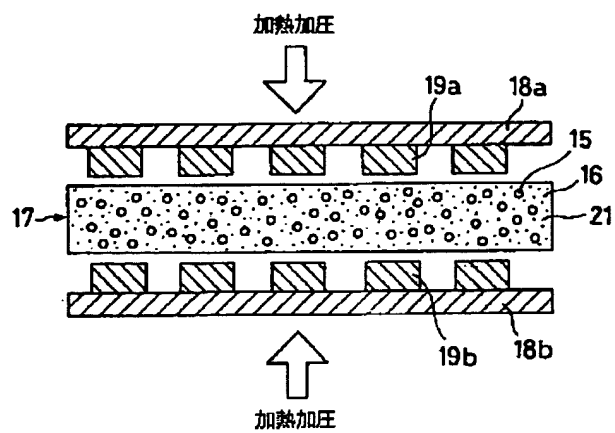
	圧着条件 180℃, 45Kg/cm <sup>2</sup> , 5秒		圧着条件 180℃, 45Kg/cm <sup>2</sup> , 30秒	
	初期導電性	経時後の導電性*	初期導電性	経時後の導電性*
実施例 2	5 Ω	30 Ω	5 Ω	5 Ω
実施例 3	5 Ω	30 Ω	5 Ω	5 Ω
実施例 4	5 Ω	30 Ω	5 Ω	5 Ω
実施例 5	5 Ω	30 Ω	5 Ω	5 Ω
実施例 6	200 Ω以上	200 Ω以上	5 Ω	10 Ω
実施例 7	200 Ω以上	200 Ω以上	10 Ω	10 Ω
実施例 8	200 Ω以上	200 Ω以上	10 Ω	10 Ω
実施例 9	200 Ω以上	200 Ω以上	20 Ω	20 Ω
実施例 10	200 Ω以上	200 Ω以上	20 Ω	20 Ω
比較例 2	1000 Ω以上	1000 Ω以上	5 Ω	1000 Ω以上

註) 「経時後の導電性\*」は、シートを40℃×30日間保管した試料を圧着し、  
 23℃×24時間後測定した導電性である。  
 導電性は、基板端部における導電性である。

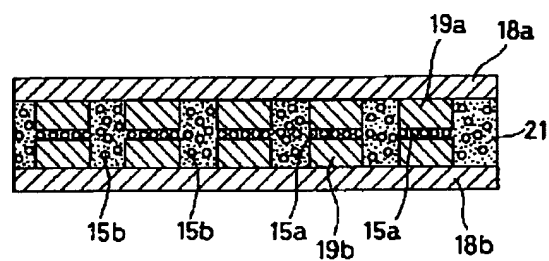
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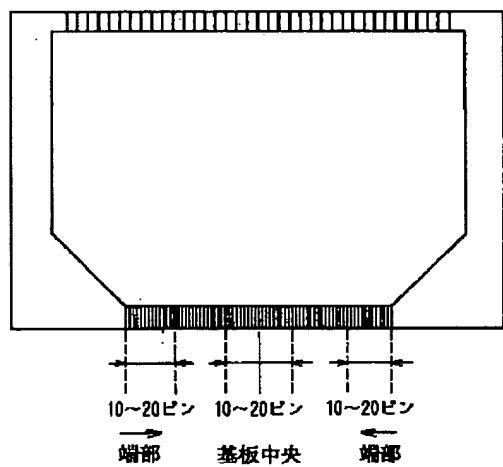
[ a ]



[ b ]



Drawing 1



Drawing 2